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STUDY OF RECRYSTALLIZATION OF BAKED ALUMINUM POWDER (SAP)

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Recently, among the heat-resisting material used for construction, the alloys of a metal system - oxide, obtained by the powder metallurgy method (Ref 4 and 5) have been used more extensively. One of the alloys used most extensively is baked aluminum powder with the oxide Al_2O_3, called SAP (Ref 5). This is a typical alloy.

The structure of alloys of the type SAP (Figure 1) constitutes a conglomerate of dispersed particles of metal oxides, relatively evenly distributed in baked metal base. In distinction from alloys hardening under dispersion, the second phase in SAP is less dispersed and is distributed more coarsely in the matrix oxide. Particles do not dissolve in aluminum, and therefore do not participate in processes occuring during thermal treatment. Furthermore, oxide particles barely coagulate down to the melting temperature of the base. This ensures high thermal stability of alloys of the type SAP. As a result at high temperatures SAP has significantly higher durability characteristics than alloys hardening under dispersion. However, at low temperatures the durability of SAP is lower (see Figure 2).

Attempts to develop alloys of type SAP on the basis of higher refractory metals than aluminum - copper, nickel, molybdenum with oxides, or other refractory compounds - are very promising. In Figure 3 is shown the dependence of long durability on test temperature for pure copper and an alloy of copper with 10%(volume) Al_2O_3 (Ref 4). Long durability of the alloy Cu plus Al_2O_3 for 1,000hrs at a temperature 350°C is 4 times greater

than for pure copper. An alloy of nickel with 10% Al_2O_3 in the temperature range of 700-980°C is significantly more stable than nickel and many complicated alloys based upon it (Ref 6). Thus, the time interval before destruction occurred at a temperature of 815°C and stress of 34.355 MN/m^2 (3.5 kG/mm^2) for the alloy Ni plus Al_2O_3 turned out to be equal 2,000hrs and for nickel only 0.2hrs.

Figure 4 shows that small quantities of dispersed inclusions of various types of oxides increase the creep resistance of molybdenum at 982°C (ref 7).

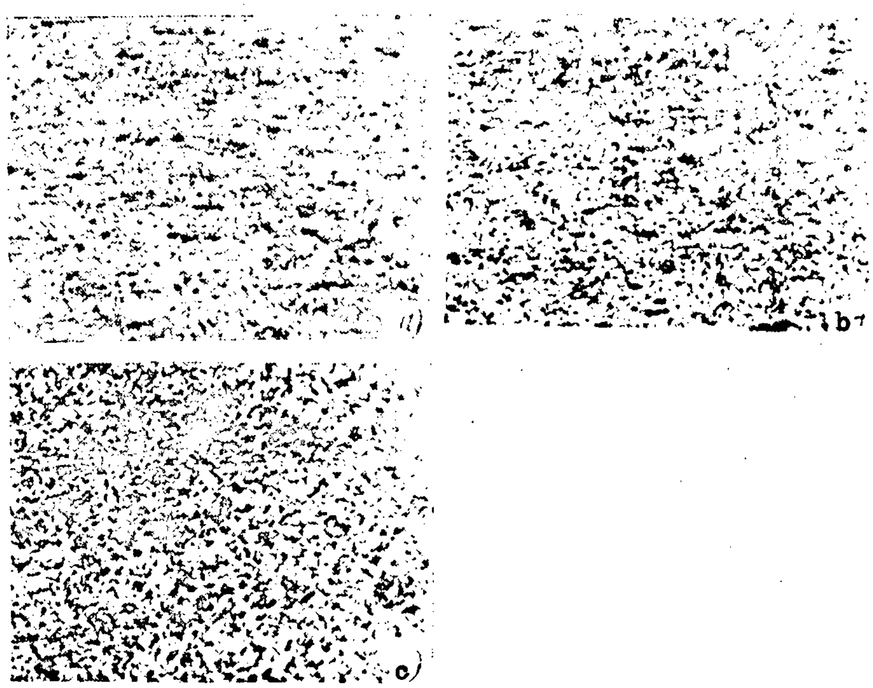

Fig 1. Microstructure of SAP after cold rolling - a); after cold rolling and annealing at a temperature of 400°C b); and after cold rolling and annealing at a temperature of 500°C c).

According to (Ref 4), alloys of the type SAP, besides high heat resistance, possess fully satisfactory resistance values to thermal shock, sensitivity to cutting, and others.

Heat resistance of alloys of the type SAP is determined by many factors. In particular, the dispersed phase must not be dissolved in a base, but should ensure the necessary cohesion with it. The heat resistance of an alloy is higher the less the dimension of particles of the dispersed phase (Figure 5) and the less the distance between them (Ref 8). Optimum dimensions of metal particles up to pressing oscillate from tenth of fractions-of a micron to several micron and of the dispersed phase - hundredths of a micron (Ref 4).

For the optimum combination of durability and plasticity properties, the amount of the dispersed phase and the uniformity of its distribution is of great value. Thus, in SAP the dispersed phase should be 10-12 vol.% (in the case Ni plus Al_2O_3 - about 10 vol.%). A uniform distribution is ensured by thorough mixing of powders before pressing.

The preparation method for alloys of the type SAP includes the operation of mixing, cold and hot pressing, sintering and pressing of half-finished product.

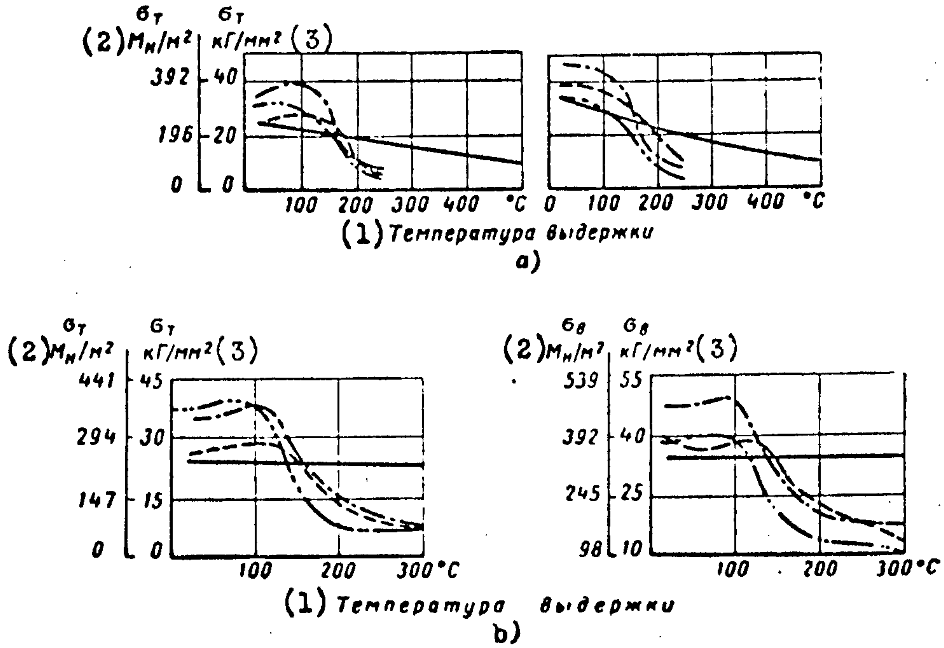

Fig 2. Short term durability of SAP and aluminum alloys in an old condition after holding them for two years at various temperatures:
——— SAP; ---- the alloy Al-Cu-Ni; — · — the alloy Al-Cu-Mg; ..... the alloy Al-Mg-Si; a) test at the holding temperature; b) test at a temperature of 20°C. 1) Holding temperature; 2) Mn/m^2; 3) kg/mm^2.

For the final pressing treatment, depending upon the form of obtained articles, rolling, stamping or pressing can be used.

For simple alloys of the type metal-oxide, the mechanism of their hardening and recrystallization during heating is still not fully clear. It is necessary to know the fine structure of the alloys, which has been studied

very little.

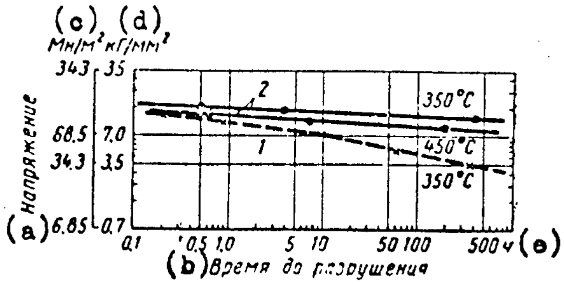

Fig 3. Prolonged durability of copper (1) and alloys Cu plus 10% Al_2O_3 (2) at various test temperatures. a) Stress; b) Time before destruction; c) Mn/m^2; d) kg/mm^2; e) hours.

Data on recrystallization of baked alloys are contradictory (Ref 4, 5). Certain authors (Ref 5), on the basis of the fact that the growth of grains in SAP is retarded by particles of a dispersed phase, draw the conclusion that SAP in general, does not recrystallize.

The following article starts with an investigation of the mechanism of recrystallization in deformed SAP.

MATERIAL AND METHOD OF INVESTIGATION

The investigation was carried out on samples, prepared from aluminum powder, containing only 4%Al_2O_3. This facilitated observation of initial stages of recrystallization. Initial ingots produced by rolling were obtained by the method of hot pressing and had a complicated axial texture, including two orientations (111) and (110). Hot rolling was done at a temperature of 450-480°C in three stages up to degree of deformation: 12.5%, 22% and 22% with two intermediate heatings to the rolling temperature. Cold rolling was produced with pressing of 65% without intermediate heatings. After deformation the samples were heated to 150-700°C for 1hr. and were cooled in air.

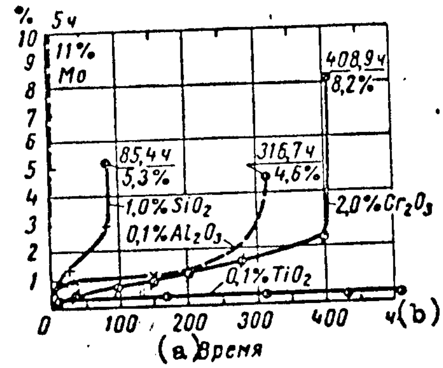

Fig 4. Creep of molybdenum and alloys Mo plus oxide at a temperature of 982°C and stress 172.5 Mn/m^2. Test in vacuum.
a) Time; b) hours.

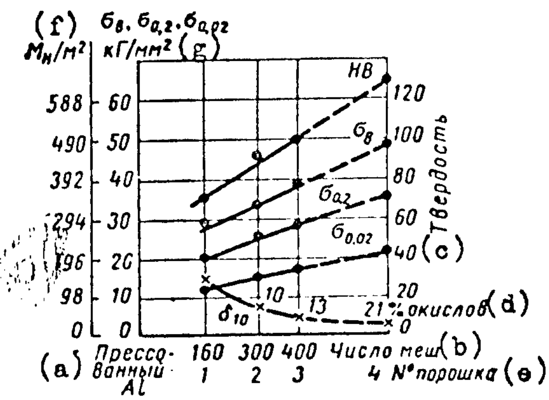

Fig 5. Influence of dispersiveness of oxidized particles on mechanical properties of alloys of the type SAP.
a) pressed; b) number of meshs; c) oxides;
d) hardness; e) number of powder; f) Mn/m^2; g) kg/mm^2.

After that, the hardness was determined and the microstructure and, in particular, the nature of the distribution of dispersed oxidized phase were studied. To detect early stages of recrystallization, X-ray diffraction analysis was used.

In works (9), (10) it was shown that the use of the usual (metallographic and roentgenographic) methods does not permit one to establish the true beginning of recrystallization in old alloys, and also in alloys with large quantity of dispersed phase, since the effect of recrystallization is here hidden by the influence of the dispersed phase.

In work (10) it was revealed that the true beginning of recrystallization appears in the length of texture maxima in X-ray photographs. This phenomenon was used in the described work.

To analyze changes in character of texture in the early stages of recrystallization, polar figures were constructed according to the X-ray photographs, taken in a chamber with the use of radiation from molybdenum anode. Maxima on the lines were analyzed (111) and (201). Samples cut out in the form of plates from the middle of the investigated pressed and cable samples. Furthermore, the usual roentgenographic method was used to determine the beginning of the intensive increase of recrystallization seeds by the appearance of point reflexes. In this case, the photographing was done on a flat film according to the reflection method with radiation from a copper anode.

RESULTS OF THE INVESTIGATION AND THEIR ANALYSIS

Cold rolling. In Figure 6 is shown the hardness dependence of cold-rolled SAP on the temperature of heating. Two temperature intervals of recrystallization are revealed: first - near 200-250°C, second - near 350-400°C.

For comparison, the same hardness measurements were performed on aluminum of the same composition as aluminum with a base of SAP. The aluminum preliminarily was subjected to the same cold deformation as was the SAP. Instead of two temperature intervals of hardness decrease, which were observed in SAP, in aluminum (curve 1 in Figure 6) only one interval near 225oC, connected with recrystallization, was observed. This temperature was somewhat higher than the temperature interval of the first weak recrystallization of SAP.

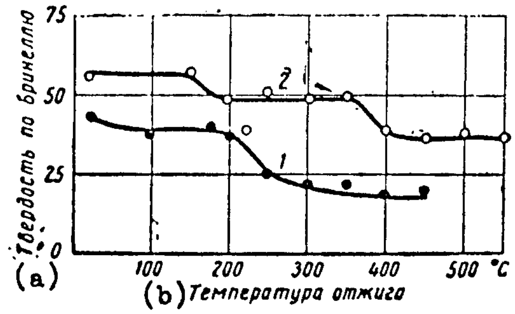

Fig 6. Hardness dependence of aluminum cold-deformed by 65% (1) and SAP (2) on annealing temperature. **a) Brinell hardness; b) annealing temperature.**

X-ray analysis revealed that heating SAP to 250°C is accompanied by a decrease in the texture scattering. Extent of texture maxima decreased (Figure 7, a and b). A more thorough investigation and analysis of the polar figures (Figure 8) helped to establish the fact that, as a result of heating to temperature 250°C, not only the texture scattering of cold-rolled SAP decreased, but the character of the texture itself was changed. In particular, orientation (111), predominant in the deformed state, disappeared.

The absence of a maximum in the center of the polar figure for planes (111) (see Figure 8) points to this. Thus during heating of cold-rolled SAP to a temperature of 200-250°C primary recrystallization occurs, but seed dimensions remain very small lines in the X-ray photograph remain solid, and recrystallization is localized in the microvolumes.

After heating to a temperature of 250°C for 1hr, on X-ray photograph is revealed weakening of texture maxima and the appearance of separate, large reflexes from large grains. Heating to 400°C strengthens this effect (Figure 7, b). After 450°C traces of texture maxima almost completely disappear, and X-ray photograph takes on a form typical for very coarse-grained samples.

Fig 7. X-ray photograph of SAP after hot pressing and subsequent cold rolling (a); after cold rolling and heating to a temperature of 250°C (b); after heating to a temperature of 400°C; (c) after hot pressing and subsequent hot rolling; (d) after hot rolling and heating to a temperature of 625°C; (e) and after hot rolling and heating to a temperature of 650°C (f).

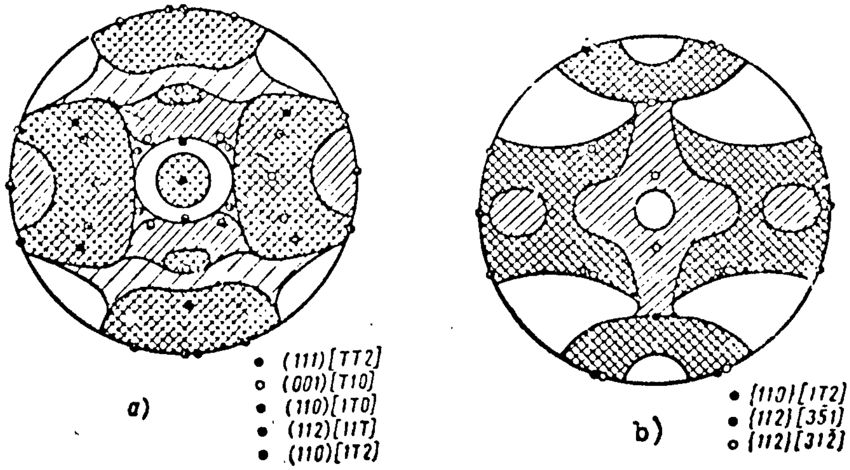

Fig 8. Polar figures (111) of the aluminum matrix of SAP after cold rolling (a); after cold rolling and subsequent heating (b).

The results of metallographic analysis were extraordinarily interesting. It was not possible to reveal the grain boundaries of aluminum matrix after their enlargement as a result of recrystallization. However, it was possible to reveal an interesting phenomenon of redistribution of oxidized particles, coinciding with the beginning of intense collective growth of recrystallization seeds.

For the structure of a cold-rolled band, the clearly-expressed, horizontal distribution of oxide part-

icles is characteristic. (see Figure 1, a). Inasmuch as Al_2O_3 will not dissolve in aluminum, it would have been possible to expect that this horizonal quality would be retained during recrystallization and would disappear only with the beginning of aluminum melting. However, it was experimentally shown that the horizontal arrangement of oxidized inclusions in the cold-rolled samples is retained during heating only to a temperature of 350°C. Roentgenographically at this temperature the beginning of intensive growth in the recrystallization seeds was detected, which also led to redistribution in the location of oxide particles in the aluminum matrix and to disturbance of the horizontal quality (see Figure 1, b). During heating to a temperature of 500-550°C horizontal traces disappear completely (Figure 1, b). This interesting fact, not noted earlier in the literature, externally represents a phenomenon, similar to the Kirkendall effect (Ref 12), but it was not caused by various partial diffusion coefficients but by redistribution of vacancies under the influence of surface tension.

B. Pines (Ref 10) noted that, under the influence of forces of surface tension, a crystallite, located on one side of an unbalanced boundary, will experience compression, but located on the other side - extension. This leads to a difference in the quantity of equilibrium vacancies on both sides of the surface. In the first crystallite, near the boundary it will be less, and in the second - larger. In view of this, the flow of vacancies from the stretched crystallite to the compressed one, or the flow of atoms in the opposite direction must be established - which appears in the collective recrystallization. The difference in the concentration of vacancies should appear in another effect. It creates hydrodynamic pressure, under the influence at which a shift in the oxide particles in the direction of stretched regions with a surplus of vacancies will occur.

It must be assumed that, for such a sharply-expressed effect from the displacement of oxidized particles to occur, the quantity of vacancies must be considerable.

Formation of the necessary quantity of vacancies in our case, obviously, is connected with the fact that, during redistribution and annihilation of dislocation in the primary recrystallization process, vacancies are released which form "atmospheres" around these dislocations. This is confirmed, for example by the diffusion acceleration by primary recrystallization (Ref 2). In this case,

such formation of free vacancies occurs at a temperature of 200-225°C. The fact that a noticeable shift of particles Al_2O_3 and an increase in recrystallized seeds are observed only at a temperature of 350-400°C, amounting to about 0.7T of aluminum, is connected, obviously, with the fact that only at this melting temperature mobility of vacancies becomes sufficient enough to permit the redistribution necessary to them.

Thus, the recrystallization course in SAP differs from the recrystallization course in pure aluminum. During cold rolling of SAP, oxide particles serve as obstacles in which dislocations accumulate. Therefore, riveting of aluminum in SAP and its heterogeneity, probably, are even more significant than in samples of pure aluminum after the same deformation. For this reason, the formation process for recrystallization seeds in cold-rolled SAP starts even at a somewhat lower temperature than in pure aluminum (200°C instead of 225°C). However, due to large quantities of undissolved inclusions, the collective growth of seeds either does not occur or is limited to local regions, free of particles Al_2O_3. In the place, where the moving boundary of a growing seed encounters an inclusion, its shift is retarded. This leads to an increase in the boundary extension and the instability of surface energy. In order to compensate for this increase in surface energy, atoms have to be reported additional energy and mobility must be transmitted to the atoms, which occurs during the described mechanism, creating a difference in the concentration of vacancies. Probably with increase in quantity Al_2O_3, the beginning of an intensive growth of seeds of recrystallization is displaced to still higher temperatures.

It is possible to assume that, with a very large quantity of highly dispersed particles, they will retard not only growth, but also actual formation of recrystallization seeds blocking dislocation and preventing their shift and annhilation. This requires experimental verification.

It is necessary to note that the influence, on the recrystallization of SAP by the formation of recrystallization seeds, is significantly weaker than the influence of the growth process of these seeds of these seeds (curve 2 on Figure 6). These data will agree well with the ideas presented in recent years by Akad. G. V. Kurdyumovym and his colleagues (Ref 2) about the leading role of structure dispersiveness during hardening. As a result of these processes, recrystallization in SAP proves to be shifted

toward higher temperatures for approximately 175-200°C as compared to aluminum.

Hot rolling. During hot rolling, the orientation of the texture axis of the initial pressed ingot is kept as a crystallographic direction, parallel to the rolling direction. Therefore, differences in the location of textured maxima for pressed and hot rolled bands is not observed. Heating of pressed and hot-rolled samples to a temperature of 600°C does not indicate the location and character of the texture maxima. After one hour of heating at a temperature of 625°C, scattering of the texture noticeably increases (Figure 7, d and f). In accordance with what has been stated above, this may be explained only by initial recrystallization, which in this case, obviously is collective.

After heating to 600°C diffusion of texture maxima is even sharper (Figure 7, e). Photography by reflection reveals the beginning of recrystallization already by the usual X-ray criteria - appearance of dots on lines of the X-ray photograph. Thus, the beginning of collective recrystallization after hot rolling occurs at significantly higher temperatures than in cold rolling. After heating to a temperature of 700°C, traces of fusion are detected in the samples and the samples themselves are deformed in furnace under action of gravity. Roentgenographic analysis shows a complete change in the sample structure and an approximation to the structure of a lithium metal.

Intense recrystallization of hot-rolled SAP, after recrystallization with heating to the beginning of the melting point, is not observed.

In hot-rolled samples also, the horizontal disturbance in the location of aluminum oxides was noted. But this occurs at much higher heating temperature than in the case of cold-rolled samples. Instead of 350-400°C, the horizontal quality in hot-rolled samples is disturbed only starting at 625°C, i.e. also near the temperature at which intense collective recrystallization begins. This once again proves that there is a connection between these two processes. Both require a sufficient quantity of vacancies and their mobility. But in distinction from cold-rolling in hot-rolled SAP, the necessary quantity of vacancies is created apparently by thermal motion, and not by the primary recrystallization process. This explains the fact

that intense collecting growth occurs at a temperature, close to the melting point.

Apparently, when aluminum is rolled at 400-450°C, its plasticity is very great. Cold hardening of an aluminum matrix (including regions, bordering on oxidized particles), and also heterogeneity of the deformation which is a necessary condition for subsequent primary recrystallization, during hot rolling is considerably less than after cold deformation. This cold hardening and the phenomena accompanying it are removed directly during deformation by means of relaxation (polygonization).

CONCLUSIONS

1. The properties of recrystallization in SAP containing 4 volume% Al_2O_3, as compared to aluminum and monophased alloys in general, and also the difference in the temperature level of recrystallization and the weakening of cold-rolled and hot-rolled SAP are studied.

2. In cold-rolled SAP with 4% Al_2O_3, deformed by 65% formation processes of recrystallization seeds and their growth, and correspondingly their influence on weakening occur at different temperatures. Seed formation begins at almost the same temperatures as in pure aluminum of the original composition - at approximately 200-225°C, while the collective growth of seeds due to "barrier" action of Al_2O_3 particles occurs only at a temperature of 350°-375°C - when the diffusion of vacancies freed during primary recrystallization becomes sufficiently intense. Collective recrystallization occurs nonuniformly and is characterized by selective growth of certain seeds. With an increase in the content of the oxidized phase, growth of recrystallization seeds should shift to still higher temperatures.

3. In hot-rolled SAP, recrystallization has only a collective character and can be observed during heating to a temperature of 625°C and above, i.e. near the melting point of aluminum, when the concentration of thermal vacancies becomes sufficiently great.

4. With the example of cold-rolled SAP, we showed that the formation of recrystallization seeds is indicated to a lesser extent by the weakening, rather than the collective growth of these seeds.

5. It is shown that the process of seed formation in textured SAP, changes the degree of scattering and the texture nature. In the case, when this process occurs at a low temperature (cold-rolled SAP), scattering of texture decreases and its character changes. When seed formation occurs at a high temperature (hot-rolled SAP), scattering is increased.

6. The interesting phenomenon of shift in the insoluble oxidized inclusions in the solid matrix under the influence of diffusion processes for the growth of recrystallization seeds was discovered. This phenomenon is similar to Kirkendall's effect, but is caused, not by various partial diffusion coefficients, but by redistribution of vacancies under the effect of surface tension.

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